#### PCT

## WORLD INTELLECTUAL PROPERTY ORGANIZATION International Bureau



101	Interna	national Bureau
INTERNATIONAL APPLICATION PUBLISI	HED U	UNDER THE PATENT COOPERATION TREATY (PCT)
(51) International Patent Classification 6:		(11) International Publication Number: WO 98/54279
C11D 3/00, 3/39	A1	1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1
		(43) International Publication Date: 3 December 1998 (03.12.98)
<ul> <li>(21) International Application Number: PCT/USC</li> <li>(22) International Filing Date: 30 May 1997 (20)</li> <li>(71) Applicant: THE PROCTER &amp; GAMBLE COLUS/US]; One Procter &amp; Gamble Plaza, Cincing 45202 (US).</li> <li>(72) Inventors: ROMANO, Nicoletta; Via Calcutta, 25, Rome (IT). TRANI, Marina; Via Gualtiero Sera 1-00136 Rome (IT). WOLFF, Anne; 4570 Boom Cincinnati, OH 45247 (US).</li> <li>(74) Agents: REED, T., David et al.; The Procter &amp; Company, 5299 Spring Grove Avenue, Cincing 45217 (US).</li> </ul>	30.05.9 MPAN nati, O I–0014 afino, 2 ner Roa Gamb	BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GE, GH, HU, IL, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, TJ, TM, TR, TT, UA, UG, UZ, VN, YU, ARIPO patent (GH, KE, LS, MW, SD, SZ, UG), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, NE, SN, TD, TG).  Published  With international search report.
(54) Title: DISINFECTING COMPOSITIONS AND PRO	CESSI	SES FOR DISINFECTING SURFACES
(57) Abstract		
	tion, of having	of a peroxygen bleach and an essential oil or an active thereof or a mixture ag been treated with said composition.
·		
·		
·		
		•

### FOR THE PURPOSES OF INFORMATION ONLY

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

1							
AL	Albania	ES	Spain	LS	Lesotho	SI	Slovenia
AM	Armenia	FI	Finland	LT	Lithuania	SK	Slovakia
AT	Austria	FR	France	LU	Luxembourg	SN	Senegal
AU	Australia	GA	Gabon	LV	Latvia	SZ	Swaziland
AZ	Azerbaijan	GB	United Kingdom	MC	Monaco	TD	Chad
BA	Bosnia and Herzegovina	GE	Georgia	MD	Republic of Moldova	TG	Togo
BB	Barbados ´	GH	Ghana	MG	Madagascar	TJ	Tajikistan
BE	Belgium	GN	Guinea	MK	The former Yugoslav	TM	Turkmenistan
BF	Burkina Faso	GR	Greece		Republic of Macedonia	TR	Turkey
BG	Bulgaria	ĦU	Hungary	ML	Mali	TT	Trinidad and Tobago
BJ	Benin	1E	Ireland	MN	Mongolia	UA	Ukraine
BR	Brazil	IL	Israel	MR	Mauritania	UG	Uganda
BY	Belarus	IS	Iceland	MW	Malawi	US	United States of America
CA	Canada	IT	Italy	MX	Mexico	UZ	Uzbekistan
CF	Central African Republic	JP	Japan	NE	Niger	VN	Viet Nam
CG	Congo	KE	Kenya	NL	Netherlands	YU	Yugoslavia
CH	Switzerland	KG	Kyrgyzstan	NO	Norway	zw	Zimbabwe
CI	Côte d'Ivoire	KP	Democratic People's	NZ	New Zealand		25255
CM	Cameroon		Republic of Korea	PL	Poland	_	
CN	China	KR	Republic of Korea	PT	Portugal		
CU	Cuba	· KZ	Kazakstan	RO	Romania		
CZ	Czech Republic	LC	Saint Lucia	RU	Russian Federation		•
DE	Germany	LI	Liechtenstein	SD	Sudan		
DK	Denmark	LK	Sri Lanka	SE	Sweden		•
EE	Estonia	LR	Liberia	SG	Singapore		

DISINFECTING COMPOSITIONS AND PROCESSES FOR DISINFECTING SURFACES

10

15

#### Technical field

The present invention relates to antimicrobial compositions which can be used to deliver long lasting disinfection on animate surfaces (e.g., human skin, mouth and the like) and inanimate surfaces including, but not limited to, hard surfaces like walls, tiles, table tops, glass, bathroom surfaces, kitchen surfaces, dishes as well as fabrics, clothes, carpets and the like.

25

30

35

20

#### Background of the invention

Antimicrobial/antibacterial compositions include materials which have the ability to disinfect. It is generally recognised that a disinfecting material greatly reduces or even eliminates the microorganisms, e.g., bacteria, existing on a surface. Typical conventional compositions although providing immediate disinfection when applied onto a surface to be disinfected are not fully satisfactory when it is also desired to prevent subsequent recontamination from microorganisms on such a surface having been first treated with said compositions. Such recontamination may happen when a surface that has been treated with an

10

antimicrobial/antibacterial composition is, later on, touched with contaminated hands or comes in contact with another source of contamination.

It is therefore an object of the present invention to provide not only effective immediate disinfection, but also effective long lasting disinfection.

This object can be met by combining a peroxygen bleach and an antimicrobial essential oil or an active thereof or a mixture thereof. Indeed, it has surprisingly been found that the incorporation, in a composition, of a peroxygen bleach and an antimicrobial essential oil, or an active thereof, provides long lasting disinfection on a surface treated therewith, even at low total level of active antimicrobial ingredients.

By combining a peroxygen bleach and an antimicrobial essential oil or an active thereof, long lasting disinfection is provided on various surfaces, i.e., animate surfaces (e.g., human skin) and inanimate surfaces. Indeed, this technology is particularly suitable for hard-surfaces applications as well as in laundry applications, e.g., as a laundry detergent or laundry additive in a so called "soaking mode", "through the wash mode", or even as a laundry pretreater in a "pretreatment mode".

An advantage of the present invention is that effective long lasting disinfection is provided on a broad range of bacterial pure strains. Indeed, the growth of Gram positive and Gram negative bacterial strains and more resistant microorganisms like fungi is reduced or even prevented on a surface when said surface has been first treated with a composition comprising a peroxygen bleach and an antimicrobial essential oil or an active thereof.

A further advantage of the present invention is that effective long lasting disinfection is provided by using ingredients which are perceived by the consumers as being safe to the surfaces treated and environmental friendly.

25

20

25

30

35

005427041 | -

Representative of the prior art is for example EP-B-288 689, which discloses a liquid for hard-surfaces comprising antimicrobial effective amounts of pine oil and at least one oil soluble organic acid. No peroxygen bleaches are disclosed.

5 US-5,403,587 discloses aqueous antimicrobial compositions which can be used to sanitise, disinfect, and clean hard-surfaces. More particularly, US-5,403,587 discloses aqueous compositions (pH 1 to 12) comprising essential oils (0.02% to 5%), which exhibit antimicrobial properties efficacy such as thyme oil, eucalyptus oil, clove oil and the like, and a solubilizing or dispersing agent sufficient to form an aqueous solution or dispersion of said essential oils in a water carrier. No peroxygen bleaches are disclosed.

None of these prior art documents discloses that by combining a peroxygen bleach with an antimicrobial essential oil or an active thereof, in a composition, long lasting disinfection is delivered to a surface having been treated therewith, even at low total levels of antimicrobial ingredients (i.e. under diluted conditions according to the present invention).

#### Summary of the invention

The present invention encompasses the use, in a composition, of a peroxygen bleach and an antimicrobial essential oil or an active thereof or mixture thereof, for long lasting disinfection on a surface treated with said composition.

#### Detailed description of the invention

A first essential ingredient of the present invention is a peroxygen bleach or a mixture thereof. Preferred peroxygen bleach is hydrogen peroxide, or a water soluble source thereof, or mixtures thereof. Hydrogen peroxide is most preferred to be used herein.

The presence of said peroxygen bleach especially hydrogen peroxide, persulfate and the like, in the compositions according to the present invention contribute to the disinfection properties of said compositions. Indeed, said peroxygen bleach

10

may attack the vital function of the micro-organism cells, for example, it may inhibit the assembling of ribosomes units within the cytoplasm of the microorganisms cells. Also said peroxygen bleach like hydrogen peroxide, is an oxidiser that generates hydroxyl free radicals which attack proteins and nucleic acids. Furthermore, the presence of said peroxygen bleach, especially hydrogen peroxide, provides strong stain removal benefits which are particularly noticeable for example in laundry and hard surfaces applications.

As used herein a hydrogen peroxide source refers to any compound which produces hydrogen peroxide when said compound is in contact with water. Suitable water-soluble sources of hydrogen peroxide for use herein include percarbonates, persilicates, persulphates such as monopersulfate, perborates and peroxyacids such as diperoxydodecandioic acid (DPDA), magnesium perphthalic acid and mixtures thereof.

In addition, other classes of peroxides can be used as an alternative to hydrogen peroxide and sources thereof or in combination with hydrogen peroxide and sources thereof. Suitable classes include dialkylperoxides, diacylperoxides, preformed percarboxylic acids, organic and inorganic peroxides and/or hydroperoxides.

- Typically, peroxygen bleach or a mixture thereof is present in the compositions according to the present invention at a level of at least 0.01% by weight of the total composition, preferably from 0.1% to 15%, and more preferably from 1% to 10%.
- A second essential ingredient of the present invention is an antimicrobial essential oil or an active thereof, or a mixture thereof.

Suitable antimicrobial essential oils to be used herein are those essential oils which exhibit antimicrobial activity. By "actives of essential oils", it is meant herein any ingredient of essential oils that exhibit antimicrobial activity. It is speculated that said antimicrobial essential oils and actives thereof act as proteins denaturing agents. Also said antimicrobial oils and actives thereof are compounds which contribute to the safety profile of a composition comprising them when it is used to disinfect any surface. A further advantage of said antimicrobial oils and actives

10

15

20

25

30

35

thereof is that they impart pleasant odor to a composition comprising them without the need of adding a perfume.

Such antimicrobial essential oils include, but are not limited to, those obtained from thyme, lemongrass, citrus, lemons, oranges, anise, clove, aniseed, pine, cinnamon, geranium, roses, mint, lavender, citronella, eucalyptus, peppermint, camphor, ajowan, sandalwood, rosmarin, vervain, fleagrass, lemongrass, ratanhiae, cedar and mixtures thereof. Preferred antimicrobial essential oils to be used herein are thyme oil, clove oil, cinnamon oil, geranium oil, eucalyptus oil, peppermint oil, citronella oil, ajowan oil, mint oil or mixtures thereof.

Actives of essential oils to be used herein include, but are not limited to, thymol (present for example in thyme, ajowan), eugenol (present for example in cinnamon and clove), menthol (present for example in mint), geraniol (present for example in geranium and rose, citronella), verbenone (present for example in vervain), eucalyptol and pinocarvone (present in eucalyptus), cedrol (present for example in cedar), anethol (present for example in anise), carvacrol, hinokitiol, berberine, ferulic acid, cinnamic acid, methyl salicylic acid, methyl salycilate, terpineol, limonene and mixtures thereof. Preferred actives of essential oils to be used herein are thymol, eugenol, verbenone, eucalyptol, terpineol, cinnamic acid, methyl salicylic acid, limonene, geraniol or mixtures thereof.

Thymol may be commercially available for example from Aldrich, eugenol may be commercially available for example from Sigma, Systems - Bioindustries (SBI) - Manheimer Inc.

Typically, the antimicrobial essential oil or active thereof or mixture thereof is present in the composition at a level of at least 0.003% by weight of the total composition, preferably from 0.006% to 10%, more preferably from 0.1% to 8% and most preferably of from 0.03% to 3%.

It has now been found that combining said antimicrobial essential oil or an active thereof or a mixture thereof with a peroxygen bleach, in a composition, delivers not only excellent immediate disinfecting properties to the surfaces treated with said composition, but also long lasting disinfecting properties. Indeed, it is speculated that peroxygen bleach and said essential oils/actives adsorb on a

10

15

surface having been treated with said composition and thus reduce or even prevent the contamination of microorganisms over time, typically up to 48 hours after the surface has been treated with said composition, thereby delivering long lasting disinfection. In other words, it is speculated that a microfilm of said active ingredients is deposited on the surface treated with said compositions allowing protection against microorganisms recontamination overtime. Advantageously, this long lasting disinfection benefits is obtained with the compositions of the present invention comprising peroxygen bleach and antimicrobial essential oils/actives even when used under highly diluted conditions, i.e., up to dilution levels of from 1:100 (composition:water).

Excellent long lasting disinfection is obtained by treating a surface with a composition comprising a peroxygen bleach and an antimicrobial essential oil or active thereof as described herein, on a variety of microorganisms, e.g., the growth of Gram positive bacteria like *Staphylococcus aureus*, and Gram negative bacteria like *Pseudomonas aeroginosa* as well as of fungi like *Candida albicans* is reduced or even prevented on a surface having been treated with said composition.

20 Long lasting disinfection properties of the compositions herein may be measured by the bactericidal activity of said compositions. A test method suitable to evaluate the long lasting bactericidal activity of a composition may be as follow: First, the surfaces (e.g. glass) to be tested are respectively treated with either a composition according to the present invention or a reference composition, e.g., 25 a negative control composed of pure water (for example by spraying the composition directly on the surface or first spraying the composition on a sponge used to clean the surface or when the composition herein is executed in the form of wipe by wiping the surface therewith). After a variable time frame (e.g. 24 hours) each surface is respectively inoculated with bacteria (10<sup>6-7</sup>cfu/slide) 30 cultured in for example TSB (Tryptone Soya Broth) and left typically from a few seconds to 2 hours before evaluating the remaining living bacteria. Then living bacteria (if any) are recovered from the surface (by touching TSA + neutraliser plates and by re-suspending the bacteria into the neutralisation broth and plating them on agar) and incubated at appropriate temperature, e.g. 37°C to let them 35 grow typically over night. Finally, a visual grading of the living bacteria is made by comparing side by side the cultures and/or dilutions thereof (e.g. 10-2 or 10-1)

resulting from the surfaces treated with the compositions according to the present invention and the reference composition.

The combination of ingredients according to the present invention, i.e., a peroxygen bleach and an antimicrobial essential oil and/or active thereof, may be formulated in a composition being either in a solid, pasty or liquid form. In the case where the compositions according to the present invention are formulated as solids, they will be mixed with an appropriate solvent, typically water, before use. In liquid form, the compositions are preferably but not necessarily formulated as aqueous compositions. Liquid compositions are preferred herein for convenience of use.

In a preferred embodiment the liquid compositions according to the present invention are aqueous compositions typically comprising from 50% to 99% by weight of the total composition of water, preferably from 70% to 99% and more preferably from 80% to 99%. These aqueous compositions preferably have a pH as is of not more than 12.0, more preferably from 1 to 10, and most preferably from 2 to 9. The pH of the compositions can be adjusted by using organic or inorganic acids, or alkalinising agents.

20

25

30

35

5

10

15

#### Optional ingredients

In a preferred embodiment of the present invention, as effective long lasting disinfection is provided with the combination of ingredients of the present invention the compositions comprising said combination do not require the addition of other antimicrobial compounds. However, in particular embodiments of the present invention, depending on the end use desired with said compositions they may further comprise, as optional ingredients, other antimicrobial compounds that further contribute to the antimicrobial/antibacterial activity of the compositions according to the present invention. Such antimicrobial ingredients include parabens like ethyl paraben, propyl paraben, methyl paraben, glutaraldehyde or mixtures thereof.

The compositions herein may further comprise a variety of other optional ingredients such as surfactants, chelants, solvents, builders, stabilisers, bleach

activators, soil suspenders, dye transfer agents, brighteners, perfumes, anti dusting agents, enzymes, dispersant, dye transfer inhibitors, pigments, perfumes, radical scavengers, pH buffers, dyes or mixtures thereof.

- The compositions according to the present invention may comprise any surfactant known to those skilled in the art including nonionic, anionic, cationic, amphoteric and/or zwitterionic surfactants. Said surfactants are suitable as they contribute to the cleaning performance of the compositions herein.
- Typically, the compositions according to the present invention comprise up to 50% by weight of the total composition of a surfactant, or mixtures thereof, preferably from 0.3 % to 30 % and more preferably from 0.4 % to 25 %.
  - Particularly suitable anionic surfactants for use herein include water-soluble salts or acids of the formula ROSO<sub>3</sub>M wherein R is preferably a C<sub>6</sub>-C<sub>24</sub> hydrocarbyl, preferably an alkyl or hydroxyalkyl having a C<sub>8</sub>-C<sub>20</sub> alkyl component, more preferably a C<sub>8</sub>-C<sub>16</sub> alkyl or hydroxyalkyl, and M is H or a cation, e.g., an alkali metal cation (e.g., sodium, potassium, lithium), or ammonium or substituted ammonium (e.g., methyl-, dimethyl-, and trimethyl ammonium cations and quaternary ammonium cations, such as tetramethyl-ammonium and dimethyl piperdinium cations and quaternary ammonium cations derived from alkylamines such as ethylamine, diethylamine, triethylamine, and mixtures thereof, and the like).
- Other suitable anionic surfactants for use herein include alkyl-diphenyl-ethersulphonates and alkyl-carboxylates. Other anionic surfactants can include salts
  (including, for example, sodium, potassium, ammonium, and substituted
  ammonium salts such as mono-, di- and triethanolamine salts) of soap, Cg-C20
  linear alkylbenzenesulfonates, C8-C22 primary or secondary alkanesulfonates,
  C8-C24 olefinsulfonates, sulfonated polycarboxylic acids prepared by sulfonation
  of the pyrolyzed product of alkaline earth metal citrates, e.g., as described in
  British patent specification No. 1,082,179, C8-C24 alkylpolyglycolethersulfates
  (containing up to 10 moles of ethylene oxide); alkyl ester sulfonates such as C1416 methyl ester sulfonates; acyl glycerol sulfonates, fatty oleyl glycerol sulfates,
  alkyl phenol ethylene oxide ether sulfates, paraffin sulfonates, alkyl phosphates,
  isethionates such as the acyl isethionates, N-acyl taurates, alkyl succinamates

15

20

and sulfosuccinates, monoesters of sulfosuccinate (especially saturated and unsaturated C<sub>12</sub>-C<sub>18</sub> monoesters) diesters of sulfosuccinate (especially saturated and unsaturated C6-C14 diesters), acyl sarcosinates, sulfates of alkylpolysaccharides such as the sulfates of alkylpolyglucoside (the nonionic nonsulfated compounds being described below), branched primary alkyl sulfates, polvethoxy carboxylates alkyl such those the formula  ${\rm RO(CH_2CH_2O)_kCH_2COO-M}^+$  wherein R is a  ${\rm C_8-C_{22}}$  alkyl, k is an integer from 0 to 10, and M is a soluble salt-forming cation. Resin acids and hydrogenated resin acids are also suitable, such as rosin, hydrogenated rosin, and resin acids and hydrogenated resin acids present in or derived from tall oil. examples are given in "Surface Active Agents and Detergents" (Vol. I and II by Schwartz, Perry and Berch). A variety of such surfactants are also generally disclosed in U.S. Patent 3,929,678, issued December 30, 1975 to Laughlin, et al. at Column 23, line 58 through Column 29, line 23.

15

20

25

30

35

10

5

Preferred anionic surfactants for use herein are the C8-C16 alkyl sulfonates, C8-C16 alkyl sulfates, C8-C16 alkyl alkoxylated sulfates (e.g., C8-C16 alkyl ethoxylated sulfates), and mixtures thereof. Such anionic surfactants are preferred herein as it has been found that they contribute to the disinfecting properties of a disinfecting composition herein. For example, C8-C16 alkyl sulfate acts by disorganizing the bacteria cell membrane, inhibiting enzymatic activities, interrupting the cellular transport and/or denaturing cellular proteins. Indeed, it is speculated that the improved disinfecting performance further associated with the addition of an anionic surfactant, especially a C8-C16 alkyl sulfonate, a C8-C16 alkyl sulfate and/or a C8-C16 alkyl alkoxylated sulfate, in a composition according to the present invention, is likely due to multiple mode of attack of said surfactant against the bacteria.

Suitable nonionic surfactants for use herein are fatty alcohol ethoxylates and/or propoxylates which are commercially available with a variety of fatty alcohol chain lengths and a variety of ethoxylation degrees. Indeed, the HLB values of such alkoxylated nonionic surfactants depend essentially on the chain length of the fatty alcohol, the nature of the alkoxylation and the degree of alkoxylation. Surfactant catalogues are available which list a number of surfactants, including nonionics, together with their respective HLB values.

Particularly suitable for use herein as nonionic surfactants are hydrophobic nonionic surfactants having an HLB (hydrophilic-lipophilic balance) below 16, preferably below 15 and more preferably below 14. Those hydrophobic nonionic surfactants have been found to provide good grease cutting properties.

5

10

15

20

25

Preferred hydrophobic nonionic surfactants for use in the compositions according to the present invention are surfactants having an HLB below 16 and being according to the formula RO- $(C_2H_4O)_{n}(C_3H_6O)_{m}H$ , wherein R is a  $C_6$  to C22 alkyl chain or a C6 to C28 alkyl benzene chain, and wherein n+m is from 0 to 20 and n is from 0 to 15 and m is from 0 to 20, preferably n+m is from 1 to 15 and, n and m are from 0.5 to 15, more preferably n+m is from 1 to 10 and, n and m are from 0 to 10. The preferred R chains for use herein are the C<sub>8</sub> to C<sub>22</sub> alkyl chains. Accordingly, suitable hydrophobic nonionic surfactants for use herein are Dobanol R 91-2.5 (HLB= 8.1; R is a mixture of C9 and C<sub>11</sub> alkyl chains, n is 2.5 and m is 0), or Lutensol  $^{\hbox{\scriptsize R}}$  TO3 (HLB=8; R is a C<sub>13</sub> alkyl chains, n is 3 and m is 0), or Lutensol R AO3 (HLB=8; R is a mixture of C<sub>13</sub> and C<sub>15</sub> alkyl chains, n is 3 and m is 0), or Tergitol R 25L3 (HLB= 7.7; R is in the range of C<sub>12</sub> to C<sub>15</sub> alkyl chain length, n is 3 and m is 0), or Dobanol R 23-3 (HLB=8.1; R is a mixture of  $C_{12}$  and  $C_{13}$  alkyl chains, n is 3 and m is 0), or Dobanol R 23-2 (HLB=6.2; R is a mixture of  $C_{12}$  and  $C_{13}$  alkyl chains, n is 2 and m is 0), or Dobanol R 45-7 (HLB=11.6; R is a mixture of  $C_{14}$  and  $C_{15}$  alkyl chains, n is 7 and m is 0) Dobanol R 23-6.5 (HLB=11.9; R is a mixture of C<sub>12</sub> and C<sub>13</sub> alkyl chains, n is 6.5 and m is 0), or Dobanol R 25-7 (HLB=12; R is a mixture of C<sub>12</sub> and C<sub>15</sub> alkyl chains, n is 7 and m is 0), or Dobanol R 91-5 (HLB=11.6; R is a mixture of Co and Cot alkyl chains, n is 5 and m is 0), or Dobanol R 91-6 (HLB=12.5; R is a mixture of C<sub>9</sub> and C<sub>11</sub> alkyl chains, n is 6 and m is 0), or Dobanol R 91-8 (HLB=13.7 ; R is a mixture of C9 and C11 alkyl chains, n is 8 and m is 0), Dobanol R 91-10 (HLB=14.2; R is a mixture of C9 to C<sub>11</sub> alkyl chains, n is 10 and m is 0), or mixtures thereof. Preferred herein are Dobanol R 91-2.5, or Lutensol R TO3, or Lutensol R AO3, or Tergitol R 25L3, or Dobanol R 23-3, or Dobanol R 23-2, or mixtures thereof. These Dobanol R surfactants are commercially available from SHELL. These LutensolR surfactants are commercially available from BASF and these Tergitol R surfactants are commercially available from UNION CARBIDE.

35

30

Other suitable surfactants also include C6-C20 conventional soaps (alkali metal salt of a C6-C20 fatty acid, preferably sodium salts).

Suitable amphoteric surfactants for use herein include amine oxides having the following formula R<sub>1</sub>R<sub>2</sub>R<sub>3</sub>NO wherein each of R1, R2 and R3 is independently a saturated substituted or unsubstituted, linear or branched hydrocarbon chains of from 1 to 30 carbon atoms. Preferred amine oxide surfactants to be used according to the present invention are amine oxides having the following formula R<sub>1</sub>R<sub>2</sub>R<sub>3</sub>NO wherein R1 is an hydrocarbon chain comprising from 1 to 30 carbon atoms, preferably from 6 to 20, more preferably from 8 to 16, most preferably from 8 to 12, and wherein R2 and R3 are independently substituted or unsubstituted, linear or branched hydrocarbon chains comprising from 1 to 4 carbon atoms, preferably from 1 to 3 carbon atoms, and more preferably are methyl groups. R1 may be a saturated substituted or unsubstituted linear or branched hydrocarbon chain.

Suitable amine oxides for use herein are for instance natural blend C8-C10 amine oxides as well as C12-C16 amine oxides commercially available from Hoechst.

20

25

5

10

15

Suitable zwitterionic surfactants for use herein contain both cationic and anionic hydrophilic groups on the same molecule at a relatively wide range of pH's. The typical cationic group is a quaternary ammonium group, although other positively charged groups like phosphonium, imidazolium and sulfonium groups can be used. The typical anionic hydrophilic groups are carboxylates and sulfonates, although other groups like sulfates, phosphonates, and the like can be used. A generic formula for some zwitterionic surfactants to be used herein is

## $R_1-N^+(R_2)(R_3)R_4X^-$

30

35

wherein R<sub>1</sub> is a hydrophobic group; R<sub>2</sub> and R<sub>3</sub> are each C<sub>1</sub>-C<sub>4</sub> alkyl, hydroxy alkyl or other substituted alkyl group which can also be joined to form ring structures with the N; R<sub>4</sub> is a moiety joining the cationic nitrogen atom to the hydrophilic group and is typically an alkylene, hydroxy alkylene, or polyalkoxy group containing from 1 to 10 carbon atoms; and X is the hydrophilic group which is preferably a carboxylate or sulfonate group. Preferred hydrophobic groups R<sub>1</sub>

10

15

20

25

are alkyl groups containing from 1 to 24, preferably less than 18, more preferably less than 16 carbon atoms. The hydrophobic group can contain unsaturation and/or substituents and/or linking groups such as aryl groups, amido groups, ester groups and the like. In general, the simple alkyl groups are preferred for cost and stability reasons.

Highly preferred zwitterionic surfactants include betaine and sulphobetaine surfactants, derivatives thereof or mixtures thereof. Said betaine or sulphobetaine surfactants are preferred herein as they help disinfection by increasing the permeability of the bacterial cell wall, thus allowing other active ingredients to enter the cell.

Furthermore, due to the mild action profile of said betaine or sulphobetaine surfactants, they are particularly suitable for the cleaning of delicate surfaces, e.g., delicate laundry or surfaces in contact with food and/or babies. Betaine and sulphobetaine surfactants are also extremely mild to the skin and/or surfaces to be treated.

Suitable betaine and sulphobetaine surfactants for use herein are the betaine/sulphobetaine and betaine-like detergents wherein the molecule contains both basic and acidic groups which form an inner salt giving the molecule both cationic and anionic hydrophilic groups over a broad range of pH values. Some common examples of these detergents are described in U.S. Pat. Nos. 2,082,275, 2,702,279 and 2,255,082, incorporated herein by reference. Preferred betaine and sulphobetaine surfactants herein are according to the formula

wherein R1 is a hydrocarbon chain containing from 1 to 24 carbon atoms, preferably from 8 to 18, more preferably from 12 to 14, wherein R2 and R3 are

10

20

25

30

35

hydrocarbon chains containing from 1 to 3 carbon atoms, preferably 1 carbon atom, wherein n is an integer from 1 to 10, preferably from 1 to 6, more preferably is 1, Y is selected from the group consisting of carboxyl and sulfonyl radicals and wherein the sum of R1, R2 and R3 hydrocarbon chains is from 14 to 24 carbon atoms, or mixtures thereof.

Examples of particularly suitable betaine surfactants include C12-C18 alkyl dimethyl betaine such as coconut-betaine and C10-C16 alkyl dimethyl betaine such as laurylbetaine. Coconutbetaine is commercially available from Seppic under the trade name of Amonyl 265®. Laurylbetaine is commercially available from Albright & Wilson under the trade name Empigen BB/L®.

Other specific zwitterionic surfactants have the generic formulas:

15 
$$R_1$$
-C(O)-N(R<sub>2</sub>)-(C(R<sub>3</sub>)<sub>2</sub>)<sub>n</sub>-N(R<sub>2</sub>)<sub>2</sub><sup>(+)</sup>-(C(R<sub>3</sub>)<sub>2</sub>)<sub>n</sub>-SO<sub>3</sub><sup>(-)</sup>  
or  $R_1$ -C(O)-N(R<sub>2</sub>)-(C(R<sub>3</sub>)<sub>2</sub>)<sub>n</sub>-N(R<sub>2</sub>)<sub>2</sub><sup>(+)</sup>-(C(R<sub>3</sub>)<sub>2</sub>)<sub>n</sub>-COO<sup>(-)</sup>

wherein each R<sub>1</sub> is a hydrocarbon, e.g. an alkyl group containing from 8 up to 20, preferably up to 18, more preferably up to 16 carbon atoms, each R<sub>2</sub> is either a hydrogen (when attached to the amido nitrogen), short chain alkyl or substituted alkyl containing from one to 4 carbon atoms, preferably groups selected from the group consisting of methyl, ethyl, propyl, hydroxy substituted ethyl or propyl and mixtures thereof, preferably methyl, each R3 is selected from the group consisting of hydrogen and hydroxy groups and each n is a number from 1 to 4, preferably from 2 to 3, more preferably 3, with no more than one hydroxy group in any  $(C(R_3)_2)$  moiety. The  $R_1$  groups can be branched and/or unsaturated. The Ro groups can also be connected to form ring structures. A surfactant of this type is C10-C14 fatty acylamidopropylene(hydroxypropylene)sulfobetaine that is available from the Sherex Company under the trade name "Varion CAS sulfobetaine"®.

The compositions herein may further comprise a chelating agent as a preferred optional ingredient. Suitable chelating agents may be any of those known to those skilled in the art such as the ones selected from the group comprising phosphonate chelating agents, aminophosphonate chelating agents, substituted heteroaromatic chelating agents, amino carboxylate chelating agents, other

carboxylate chelating agents, polyfunctionally-substituted aromatic chelating agents, biodegradable chelating agents like ethylene diamine N,N'- disuccinic acid, or mixtures thereof.

Suitable phosphonate chelating agents to be used herein include etidronic acid (1-hydroxyethylene-diphosphonic acid (HEDP)), and/or alkali metal ethane 1-hydroxydiphosphonates.

Suitable amino phosphonate chelating agents to be used herein include amino alkylene poly (alkylene phosphonates), nitrilotris(methylene)triphosphonates, ethylene diamine tetra methylene phosphonates, and/or diethylene triamine penta methylene phosphonates. Preferred aminophosphonate chelating agents to be used herein are diethylene triamine penta methylene phosphonates.

These phosphonate/amino phosphonate chelating agents may be present either in their acid form or as salts of different cations on some or all of their acid functionalities. Such phosphonate/amino phosphonate chelating agents are commercially available from Monsanto under the trade name DEQUEST®.

20 Substituted heteroaromatic chelating agents to be used herein include hydroxypiridine-N-oxide or a derivative thereof.

Suitable hydroxy pyridine N-oxides and derivatives thereof to be used according to the present invention are according to the following formula:

wherein X is nitrogen, Y is one of the following groups oxygen, -CHO, -OH, - (CH2)n-COOH, wherein n is an integer of from 0 to 20, preferably of from 0 to 10 and more preferably is 0, and wherein Y is preferably oxygen. Accordingly particularly preferred hydroxy pyridine N-oxides and derivatives thereof to be used herein is 2-hydroxy pyridine N-oxide. Hydroxy pyridine N-oxides and derivatives thereof may be commercially available from Sigma.

Polyfunctionally-substituted aromatic chelating agents may also be useful in the compositions herein. See U.S. patent 3,812,044, issued May 21, 1974, to Connor et al. Preferred compounds of this type in acid form are dihydroxydisulfobenzenes such as 1,2-dihydroxy -3,5-disulfobenzene.

5

10

15

20

A preferred biodegradable chelating agent for use herein is ethylene diamine N,N'-disuccinic acid, or alkali metal, or alkaline earth, ammonium or substitutes ammonium salts thereof or mixtures thereof. Ethylenediamine N,N'- disuccinic acids, especially the (S,S) isomer have been extensively described in US patent 4, 704, 233, November 3, 1987 to Hartman and Perkins. Ethylenediamine N,N'-disuccinic acid is, for instance, commercially available under the tradename ssEDDS® from Palmer Research Laboratories. Ethylene diamine N,N'- disuccinic acid is particularly suitable to be used in the compositions of the present invention.

Suitable amino carboxylate chelating agents useful herein include ethylene diamine tetra acetates, diethylene triamine pentaacetates, diethylene triamine pentoacetate (DTPA), N-hydroxyethylethylenediamine triacetates, nitrilotriacetates, ethylenediamine tetraproprionates, triethylenetetraaminehexaacetates, ethanoldiglycines, propylene diamine tetracetic acid (PDTA) and methyl glycine di-acetic acid (MGDA), both in their acid form, or in their alkalimetal, ammonium, and substituted ammonium salt forms. Particularly suitable to be used herein are diethylene triamine penta acetic acid (DTPA), propylene diamine tetracetic acid (PDTA) which is, for instance, commercially available from BASF under the trade name Trilon FS® and methyl glycine di-acetic acid (MGDA).

25

30

35

Further carboxylate chelating agents to be used herein includes malonic acid, salicylic acid, glycine, aspartic acid, glutamic acid, or mixtures thereof.

Typically, the compositions according to the present invention comprise up to 5% by weight of the total composition of a chelating agent, or mixtures thereof, preferably from 0.01% to 3% by weight and more preferably from 0.01% to 1.5%.

The compositions herein may comprise a radical scavenger as another optional ingredient. Suitable radical scavengers for use herein include the well-known substituted mono and di hydroxy benzenes and derivatives thereof, alkyl- and aryl carboxylates and mixtures thereof. Preferred radical scavengers for use herein

10

15

20

25

30

include di-tert-butyl hydroxy toluene (BHT), p-hydroxy-toluene, hydroquinone di-tert-butyl hydroquinone (DTBHQ), mono-tert-butyl hydroguinone (MTBHQ), tert-butyl-hydroxy anysole (BHA), p-hydroxy-anysol, benzoic acid, 2,5dihydroxy benzoic acid, 2,5-dihydroxyterephtalic acid, toluic acid, catechol, t-butyl catechol, 4-allyl-catechol, 4-acetyl catechol, 2-methoxy-phenol, 2-ethoxy-phenol, 2-methoxy-4-(2-propenyl)phenol, 3,4-dihydroxy benzaldehvde. 2.3-dihydroxy benzaldehyde, benzylamine, 1,1,3-tris(2-methyl-4-hydroxy-5-t-butylphenyl) butane, tert-butyl-hydroxy-anyline, p-hydroxy anyline as well as n-propyl-gallate. Highly preferred for use herein are di-tert-butyl hydroxy toluene, which is for example commercially available from SHELL under the trade name IONOL CP® and/or tert-butyl-hydroxy anysole. These radical scavengers further contribute to the stability of the peroxygen bleach-containing compositions herein.

Typically, the compositions according to the present invention comprise up to 5% by weight of the total composition of a radical scavenger, or mixtures thereof, preferably from 0.002% to 1.5% by weight and more preferably from 0.002% to 1%.

The compositions herein may comprise as a preferred optional ingredient a solvent or mixtures thereof. When used, solvents will, advantageously, give an enhanced cleaning to the compositions herein. Suitable solvents for incorporation in the compositions according to the present invention include propylene glycol derivatives such as n-butoxypropanol or n-butoxypropoxypropanol, water-soluble CARBITOL® solvents or water-soluble CELLOSOLVE® solvents. Water-soluble CARBITOL® solvents are compounds of the 2-(2-alkoxyethoxy)ethanol class wherein the alkoxy group is derived from ethyl, propyl or butyl. A preferred watersoluble carbitol is 2-(2-butoxyethoxy)ethanol also known as butyl carbitol. Watersoluble CELLOSOLVE® solvents are compounds of the 2-alkoxyethoxyethanol class, with 2-butoxyethoxyethanol being preferred. Other suitable solvents are benzyl alcohol, methanol, ethanol, isopropyl alcohol and diols such as 2-ethyl-1,3-hexanediol and 2,2,4-trimethyl-1,3-pentanediol and mixture thereof. Preferred solvents for use herein are n-butoxypropoxypropanol, butyl carbitol®, benzyl alcohol, isopropanol and mixtures thereof. Most preferred solvents for use herein are butyl carbitol®, benzyl alcohol and/or isopropanol.

35

The solvents may typically be present within the compositions according to the invention at a level up to 15% by weight, preferably from 2% to 7% by weight of the composition.

In the embodiment of the present invention wherein the compositions are formulated in the alkaline pH range, typically from 7.5 to 12, the compositions according to the present invention may further comprise a pH buffer or a mixture thereof, i.e. a system composed of a compound or a combination of compounds, whose pH changes only slightly when a strong acid or base is added.

10

5

Suitable pH buffers for use herein include borate pH buffer, phosphonate, silicate and mixtures thereof. Suitable borate pH buffers for use herein include alkali metal salts of borates and alkyl borates and mixtures thereof. Suitable borate pH buffers to be used herein are alkali metal salts of borate, metaborate, tetraborate, 15 octoborate, pentaborate, dodecaboron, borontrifluoride and/or alkyl borate containing from 1 to 12 carbon atoms, and preferably from 1 to 4. Suitable alkyl borate includes methyl borate, ethyl borate and propyl borate. Particularly preferred herein are the alkali metal salts of metaborate (e.g. sodium metaborate), tetraborate (e.g., sodium tetraborate decahydrate) or mixtures thereof.

Boron salts like sodium metaborate and sodium tetraborate are commercially available from Borax and Societa Chimica Larderello under the trade name sodium metaborate® and Borax®.

25

20

Typically, the compositions according to the present invention may comprise up to 15% by weight of the total composition of a pH buffer, or mixtures thereof, preferably from 0.01% to 10%, more preferably from 0.01% to 5% and most preferably from 0.1% to 3%.

30

35

#### Packaging form of the compositions:

The compositions herein may be packaged in a variety of suitable detergent packaging known to those skilled in the art. The liquid compositions herein may desirably be packaged in manually operated spray-type dispensing containers, which are usually made of synthetic organic polymeric plastic materials, and

preferably in a trigger spray dispenser or pump spray dispenser. Said spray-type dispensers allow to uniformly applied to a relatively large area of a surface to be treated the liquid disinfecting compositions of the present invention. Such spray-type dispensers are particularly suitable to treat vertical surfaces.

5

10

15

20

25

30

Suitable spray-type dispensers to be used according to the present invention include manually operated foam trigger-type dispensers sold for example by Specialty Packaging Products, Inc. or Continental Sprayers, Inc. These types of dispensers are disclosed, for instance, in US-4,701,311 to Dunnining et al. and US-4,646,973 and US-4,538,745 both to Focarracci. Particularly preferred to be used herein are spray-type dispensers such as T 8500® or T 8900® commercially available from Continental Spray International or T 8100® commercially available from Canyon, Northern Ireland. In such a dispenser the liquid composition is divided in fine liquid droplets resulting in a spray that is directed onto the surface to be treated. Indeed, in such a spray-type dispenser the composition contained in the body of said dispenser is directed through the spray-type dispenser head via energy communicated to a pumping mechanism by the user as said user activates said pumping mechanism. More particularly, in said spray-type dispenser head the composition is forced against an obstacle, e.g. a grid or a cone or the like, thereby providing shocks to help atomise the liquid composition, i.e. to help the formation of liquid droplets.

The compositions of the present invention may also be executed in the form of wipes. By "wipes", it is meant herein disposable towels, e.g. paper towels, impregnated with a liquid composition according to the present invention. Preferably said wipes are packaged in a plastic box. The advantage of this execution is a faster usage of a disinfecting composition by the user, this even outside the house, i.e. there is no need to pour the liquid compositions according to the present invention on the surfaces to be treated and to dry it out with a cloth. In other words, wipes allow immediate and long lasting disinfection of the surfaces in one step.

The compositions according to the present invention are used to treat a surface thereby delivering long lasting disinfection to said surface. By "treat", it is meant

herein applying the composition to the surface in its neat (in the preferred embodiment herein wherein the composition is liquid) or diluted form.

By "diluted form", it is meant herein that the compositions to be used, being either in a liquid or a solid form, is diluted by the user typically with water up to 100 times their weight of water, preferably into 80 to 40 times their weight of water and more preferably 60 to 40, before being applied to the surface to treat.

By "surface", it is meant herein any surface including animate surface like human skin, mouth, teeth, and inanimate surfaces. Inanimate surfaces include, but are not limited to, hard-surfaces typically found in houses like kitchens, bathrooms, or in car interiors, e.g., tiles, walls, floors, chrome, glass, smooth vinyl, any plastic, plastified wood, table top, sinks, cooker tops, dishes, sanitary fittings such as sinks, showers, shower curtains, wash basins, WCs and the like, as well as fabrics including clothes, curtains, drapes, bed linens, bath linens, table cloths, sleeping bags, tents, upholstered furniture and the like, and carpets. Inanimate surfaces also include household appliances including, but not limited to, refrigerators, freezers, washing machines, automatic dryers, ovens, microwave ovens, dishwashers and so on.

The present invention will be further illustrated by the following examples.

20

5

10

15

#### Examples

The following compositions were made by mixing the listed ingredients in the listed proportions (weight % unless otherwise specified).

	Compositions % by weight	I	İ	ili	IV	V	VI
5	Hydrogen peroxide	7.0	2.0	1.0	2.0	4.0	3.0
	Thyme oil		0.05			0.5	
	Clove oil	0.5			0.05		0.1
	Geranium oil		0.07				
	Eucalyptus oil	0.2		0.02	0.01		0.2
10	Geraniol			0.5	<u></u>	0.5	0.1
	Alkyl sulphate	4	3	10	1	4	0.5
	DETPMP	0.1	0.15	0.15	0.1	0.15	0.2
	Water and minors H2SO4 up to pH 4			up	to 100%-	***********	

	Compositions % by weight	VII	VIII	IX .	X	XI	XII
20	•						
	Hydrogen peroxide	7.0	2.0	5.0	1.0	4.0	1.5
	Thyme oil		0.4	-		0.5	
	Clove oil	0.5			0.2		0.1
	Geranium oil		0.2				
25	Eucalyptus oil	0.2		0.2	0.2		0.2
	Geraniol			0.15		0.005	0.1
	Betaine	1.5	0.1		0.03		0.1
	Amine oxide	3	1.2	1.0	1		1.0
٠	Alkyl sulphate			1.5		4	
30	HEDP	0.1			0.02		0.1
	BHT			0.1			0.05
	DTPA	0.1		0.15	0.1	0.15	
	DETPMP		0.15				0.2
•	Tetraborate	•	, <del></del>	8.0			1.0
35	Water and minors			up	to 100%-		
•	PH	4	4	9	4	4	8

- HEDP is 1-hydroxyethylene-diphosphonic acid
- DETPMP is diethylene triamine penta methylene phosphonate
  DTPA is diethylene triamine pentacetate
  Alkyl sulfate is a C10 alkyl sulphate
  Betaine is coco alkyldimethyl betaine available from Albright & Wilson under the trade name of Empigen BB/L®.
- 10 Amine oxide is N-decyldimethyl amine oxide

The compositions of the examples above are according to the present invention, they may be used neat or diluted, e.g., at a dilution level of 1:50 or of 1:25 (composition:water), to provide effective long lasting disinfecting performance on

15 the surface treated.

#### Claims

- 1. The use, in a composition, of a peroxygen bleach and an antimicrobial essential oil or an active thereof or a mixture thereof, for long lasting disinfection on a surface treated with said composition.
- 2. The use according to claim 1 wherein said antimicrobial essential oil or an active thereof or a mixture thereof is present in said composition at a level of at least 0.003% by weight of the total composition, preferably from 0.006% to 10%, more preferably from 0.1% to 8% and most preferably from 0.03% to 3%.
- 3. The use according to any of the preceding claims wherein said antimicrobial essential oil is selected from the group consisting of thyme oil, lemongrass oil, citrus oil, lemon oil, orange oil, ajowan oil, anise oil, clove oil, aniseed oil, cinnamon oil, geranium oil, rose oil, lavender oil, citronella oil, eucalyptus oil, peppermint oil, mint oil, camphor oil, sandalwood oil, cedar oil, rosmarin oil, pine oil, vervain oil, fleagrass oil, lemongrass oil, ratanhiae oil and mixtures thereof, and preferably is thyme oil, ajowan oil, citronella oil, clove oil, cinnamon oil, geranium oil, eucalyptus oil, peppermint oil, mint oil or a mixture thereof, and/or wherein said active of essential oil is selected from the group consisting of thymol, eugenol, menthol, carvacrol, verbenone, eucalyptol, cedrol, anethol, pinocarvone, geraniol, hinokitiol, berberine, ferulic acid, cinnamic acid, methyl salicylic acid, methyl salicylate, terpineol, limonene and mixtures thereof, and preferably is thymol, eugenol, verbenone, eucalyptol, terpineol, cinnamic acid, methyl salicylic acid, limonene, geraniol or a mixture thereof.
- 4. The use according to any of the preceding claims wherein said peroxygen bleach or a mixture thereof is present in said composition at a level of at least 0.01% by weight of the total composition, preferably from 0.1% to 15%, and more preferably from 1% to 10%.
- 5. The use according to any of the preceding claims wherein said p roxygen bleach is hydrogen peroxide and/or a water soluble source thereof typically selected from the group consisting of percarbonate, persilicate,

persulphate, perborate, peroxyacid, dialkylperoxide, diacylperoxide, preformed percarboxylic acid, organic and inorganic peroxides, organic and inorganic hydroperoxides and mixtures thereof, more preferably is hydrogen peroxide.

- 6. The use according to any of the preceding claims wherein said composition further comprises a surfactant up to a level of 50% by weight of the total composition, typically selected from the group consisting of anionic surfactants, nonionic surfactants, cationic surfactants, amphoteric surfactants, zwitterionic surfactants and mixtures thereof.
- 7. The use according to claim 6 wherein said surfactant is an amphoteric surfactant, preferably an amine oxide and/or a zwitterionic surfactant, preferably a betaine or sulfobetaine surfactant, and/or an anionic surfactant, preferably a C8-C16 alkyl sulfonate, C8-C16 alkyl sulfate and/or C8-C16 alkyl alkoxylated sulfate.
- 8. The use according to any of the preceding claims wherein said composition further comprises at least an ingredient selected from the group consisting of chelants, solvents, builders, stabilisers, bleach activators, soil suspenders, dye transfer agents, brighteners, perfumes, anti dusting agents, enzymes, dispersant, dye transfer inhibitors, pigments, perfumes, radical scavengers, dyes, and mixtures thereof.
- 9. The use according to any of the preceding claims wherein said composition is a liquid composition, preferably an aqueous composition having a pH of not more than 12, more preferably from 1 to 10 and most preferably from 2 to 9.
- 10. The use according to any of the preceding claims wherein said composition further comprises a pH buffer or a mixture thereof, preferably a borate pH buffer, more preferably an alkali metal salt of borate, metaborate, tetraborate, octoborate, pentaborate, dodecaboron, borontrifluoride, or an alkyl borate containing from 1 to 12 carbon atoms, or a mixture thereof, up to a level of 15% by weight of the total composition, preferably from 0.01% to 10% and more preferably from 0.1% to 3%.

# INTERNAT NAL SEARCH REPORT

I :atio..ál Application No PCT/US 97/09260

		***************************************		
A. CLASSIFICATION OF SUBJECT MATTER IPC 6 C11D3/00 C11D3/39				
According to International Patent Classification(IPC) or to both national classi	fication and IPC			
3. FIELDS SEARCHED				
Minimum documentation searched (classification system followed by classification $6000000000000000000000000000000000000$	ation symbols)			
Occumentation searched other than minimumdocumentation to the extent tha	t such documents are include	d in the fields searched		
Electronic data base consulted during the international search (name of data .	base and, where practical, se	arch terms used)		
DOCUMENTS CONSIDERED TO BE RELEVANT				
Category ° Citation of document, with indication, where appropriate, of the i	elevant passages	Relevant to claim No.		
WO 92 18091 A (PROCTER & GAMBLE October 1992 see page 13, line 28 - page 14,	,	1-9		
see claims 1-5  ( EP 0 252 278 A (HENKEL KGAA) 13	January	1-6,8-10		
1988 see page 9; examples 4,5,8-16				
see claims 1,37 see page 7, line 43 - line 57 see page 6, line 5 - line 10				
WO 88 00795 A (GARCIN FRANCOISE February 1988 see page 3, line 15 - page 4, 1 see page 2, line 21 - line 23		1-6,8		
Further documents are listed in the continuation of box C.	χ Patent family mer	mbers are listed in annex.		
Special categories of cited documents :		ned after the international filing date		
A* document defining the general state of the art which is not considered to be of particular relevance  = earlier document but published on or after the international	cited to understand the invention	ot in conflict with the application but he principle or theory underlying the		
filing date L" document which may throw doubts on priority claim(s) or which is cited to establish the publicationdate of another	cannot be considered involve an inventive of	r relevance; the claimed invention d novel or cannot be considered to step when the document is taken alone r relevance; the claimed invention		
citation or other special reason (as specified) O" document referring to an oral disclosure, use, exhibition or	cannot be considered document is combine	d to involve an inventive step when the ad with one or more other such docu-		
other means  P" document published prior to the international filing date but later than the priority date claimed	ments, such combina in the art. "&" document member of	ation being obvious to a person skilled · the same patent family		
ate of the actual completion of theinternational search	Date of mailing of the	international search report		
21 January 1998	30/01/199	98		
lame and mailing address of the ISA European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk	Authorized officer			
NL - 2200 NV NISW  K Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Richards	, M .		

Form PCT/ISA/210 (second sheet) (July 1992)

## INTERNA DNAL SEARCH REPORT

information on patent family members

Ir \_tional Application No PCT/US 97/09260

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
WO 9218091 A	29-10-92	AU 1890292 A	17-11-92
EP 0252278 A	13-01-88	DE 3702983 A CA 1277899 A JP 2109426 C JP 8018939 B JP 62292709 A US 4900721 A	10-12-87 18-12-90 21-11-96 28-02-96 19-12-87 13-02-90
WO 8800795 A	11-02-88	FR 2601850 A DE 3781885 A EP 0280697 A	29-01-88 29-10-92 07-09-88